

Crossover from Luttinger liquid to Coulomb blockade regime in carbon nanotubes

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We develop a theoretical approach to the low-energy properties of 1D electron systems aimed to encompass the mixed features of Luttinger liquid and Coulomb blockade behavior observed in the crossover between the two regimes. For this aim we extend the Luttinger liquid description by incorporating the effects of a discrete single-particle spectrum. The intermediate regime is characterized by a power-law behavior of the conductance, but with an exponent oscillating with the gate voltage, in agreement with recent experimental observations. Our construction also accounts naturally for the existence of a crossover in the zero-bias conductance, mediating between two temperature ranges where the power-law behavior is preserved but with different exponent.

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The recent progress in nanotechnology has allowed for a detailed study of the transport properties of one-dimensional (1D) electron systems. The discovery of carbon nanotubes (CNs) in 1991[1], as a by-product of fullerene production, has opened a new field of research in mesoscopic physics, especially because of their potential application to nanoelectronic devices[2].

CN are rolled up sheets of graphite forming tubes that are only nanometers in diameter, with metallic or semiconducting properties depending on the roll-up direction of the sheet. Regarding the metallic CNs, their transport properties are dictated by the low dimensionality of the system, which gives rise to strong electronic correlations. Thus, the effects of the Coulomb interaction become very important in CNs, and the way they manifest themselves depends mainly on the size of the system, the temperature, and the quality of the contacts used in experiments.

When the contacts are not highly transparent, the measurements of the conductance and the differential conductivity reflect the strong Coulomb repulsion in CNs. For temperatures that are typically below 1 K, the zero-bias conductance shows oscillations characteristic of the so-called Coulomb blockade regime[3]. The gate voltage between two peaks is related to the energy required to overcome the Coulomb repulsion when adding an electron between the barriers created by the contacts. In general, the intensity plots in terms of the bias and gate voltages show in that regime a sequence of diamonds where the electron tunneling is suppressed, with the size of the diamonds giving a measure of the energy needed to add an electron to the system[4].

For higher values of the temperature, such that the thermal energy is much larger than the level spacing, the transport measurements reflect instead the many-body properties of the system. The conductance shows then a different kind of suppression, which arises from the strongly correlated character of the electron liquid[5]. In a 1D system, the electron quasiparticles are unstable under the slightest interaction and the low-energy excitations take the form of charge or spin density fluctuations.

In the so-called Luttinger liquid regime, the absence of electron quasiparticles gives rise to a power-law suppression of the tunneling density of states (TDOS) near the Fermi level, $n(\varepsilon) \sim \varepsilon^\alpha$, which translates in turn into a similar behavior of quantities like the zero-bias conductance as a function of the temperature, $G(T) \sim T^\alpha$.

Such kind of signatures of Luttinger liquid behavior have been observed in single-walled[6] as well as in multi-walled nanotubes (MWNTs)[7]. It has been shown that the zero-bias conductance and the differential conductivity follow closely a power-law behavior as functions of the temperature and the bias voltage, respectively. In the case of the zero-bias conductance, such power-law dependence holds over a wide range of temperatures, down to values where the discrete character of the spectrum starts to become manifest. It is observed then the transition to the Coulomb blockade regime, where the suppression of the conductance arises from the single-electron tunneling mentioned above.

At this point, it would be desirable to have a theoretical description of the crossover as the temperature decreases from the Luttinger liquid regime to the Coulomb blockade regime. There have been experiments like that reported in Ref. 8 in which the intermediate regime has been explored, measuring the zero-bias conductance at temperatures where the thermal energy becomes comparable to the level spacing in the discrete spectrum. The results reported in that reference provide evidence for the power-law behavior of the TDOS in MWNTs, but with an unusual dependence of the exponent α on the gate voltage. Clear oscillations have been observed in that plot, with a characteristic scale of the order of 1 V. Another remarkable observation has been the existence of an inflection point in the log-log plot of the conductance versus temperature, for gate voltages corresponding to peak values in the α exponent[8].

The aim of the present paper is to provide a framework that may encompass the mixed features of Luttinger liquid and Coulomb blockade behavior observed in the crossover between the two regimes. For that purpose we

will rely on the 1D many-body approach[9], incorporating at the same time the discrete character of the spectrum which arises from the finite size of the system. Thus we will see that the polarizability, which is proportional to the single-particle density of states, is modulated at sufficiently low temperature due to the peaks building the density of states in the discrete spectrum. Such a modulation is translated to the scaling behavior of the quasi-particle weight, giving rise to oscillations in the exponent of the TDOS as the Fermi level of the system is shifted. We will show that the period estimated for the oscillations is in agreement with the experimental observations, and that the proposed framework is able to account for the features seen in the measurements of the zero-bias conductance reported in Ref. 8.

In our approach, a central role in distinguishing the Coulomb blockade from the Luttinger liquid regime is played by the energy distribution function $n(\varepsilon; \varepsilon_F)$. At low temperatures, that function has peaks that reflect the level spacing $\Delta\varepsilon$ arising from the level quantization in a finite nanotube. We can model each peak with a normalized function $f(\varepsilon, n)$, with a shape depending on the temperature T

$$f(\varepsilon, n) = \frac{1}{k_B T \cosh^2\left(\frac{\varepsilon - n\Delta\varepsilon}{k_B T}\right)}. \quad (1)$$

The energy distribution function is cutoff by the Fermi energy ε_F , as it is given by

$$n(\varepsilon; \varepsilon_F) = \sum_{n=0}^{\varepsilon_F/\Delta\varepsilon} f(\varepsilon, n). \quad (2)$$

At sufficiently high temperatures, we can neglect the level spacing and calculate the energy distribution function by integrating

$$\begin{aligned} n(\varepsilon; \varepsilon_F) &\approx \int_{-\infty}^{\varepsilon_F/\Delta\varepsilon - 1/2} dy f(\varepsilon, y) \\ &= \frac{1}{1 + e^{(\varepsilon - \varepsilon_F + \Delta\varepsilon/2)/k_B T}}. \end{aligned} \quad (3)$$

The last formula gives the usual Fermi-Dirac distribution. We realize however that the energy distribution function has in general a more structured shape, that depends on the ratio between the level spacing $\Delta\varepsilon$ and the thermal energy $k_B T$ as illustrated in Fig. 1.

The structure of peaks in the energy distribution function affects several objects in the many-body theory. This is the case of the one-loop polarization $\Pi^{(0)}(q, \omega_q)$, which is given in terms of the electron propagator $G^{(0)}(k, \omega_k)$ by

$$\Pi^{(0)}(q, \omega_q) = \int \frac{dk}{2\pi} \int \frac{d\omega_k}{2\pi} G^{(0)}(k+q, \omega_k + \omega_q) G^{(0)}(k, \omega_k). \quad (4)$$

This object counts the number of particle-hole excitations that can be built with momentum q across the

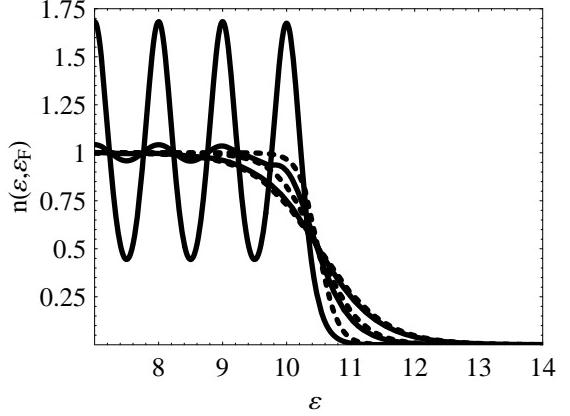


FIG. 1: Plot of the energy distribution function $n(\varepsilon; \varepsilon_F)$ for different values of the temperature corresponding to $k_B T / \Delta\varepsilon = 0.25, 0.6, 1.0$. The solid lines have been obtained from Eq. (2) and the dashed lines by using the Fermi-Dirac distribution (3).

Fermi level. Thus, in the case of a model with linear dispersion $\varepsilon(k) \approx v_F k$, it becomes proportional at zero temperature to

$$\int_{-\infty}^{\infty} dk \theta(k_F - k) \theta(q + k - k_F) = q. \quad (5)$$

However, in the model with a distribution function given by (2), the peaks give rise to a factor in the integration that reflects the discrete structure

$$\int_{-\infty}^{\infty} dk n(v_F k; \varepsilon_F) (1 - n(v_F(q+k); \varepsilon_F)) = q N_T(\varepsilon_F). \quad (6)$$

The usual form of the 1D polarization is modified then by the presence of the last factor in (6):

$$\Pi^{(0)}(q, \omega_q) = \frac{2}{\pi} N_s N_T(\varepsilon_F) \frac{v_F q^2}{\omega_q^2 - v_F^2 q^2} \quad (7)$$

with the factor N_s standing for the number of subbands crossing the Fermi level. The factor $N_T(\varepsilon_F)$ is itself a periodic function of ε_F , since the position of the Fermi energy fixes the portion of the last peak that has to be integrated in the spectrum, in order to obtain the polarizability.

We can incorporate the polarization (7) in the RPA evaluation of the electron self-energy $\Sigma(k, \omega_k)$. This can be expressed as

$$\begin{aligned} \Sigma(k, \omega_k) &= i \int_{-\Lambda/v_F}^{\Lambda/v_F} \frac{dq}{2\pi} \int_{-\infty}^{\infty} \frac{d\omega_q}{2\pi} G^{(0)}(k-q, \omega_k - \omega_q) \\ &\quad \times \frac{V(q)}{1 - V(q) \Pi^{(0)}(q, \omega_q)}, \end{aligned} \quad (8)$$

where $V(q)$ stands for the 1D Coulomb potential in momentum space. The high-energy cutoff Λ comes from the

existence of a microscopic short-distance scale in the electron system and, for our purposes, it provides a way of having under control the scale dependence of the different observables[9].

The RPA scheme allows for an accurate description of the behavior of the quasiparticle weight at low energies, since it gives the exact result for the anomalous dimension of the electron field in the 1D system[10]. Thus, the self-energy turns out to have terms linear in ω_k that depend logarithmically on the cutoff Λ . This is the signal of the energy dependence of the scale $Z^{1/2}$ of the electron wavefunction. The dressed electron propagator $G(k, \omega_k)$ is given by

$$\begin{aligned} \frac{1}{G} &= \frac{1}{G^{(0)}} - \Sigma(k, \omega_k) \\ &\approx Z^{-1}(\Lambda)(\omega_k - v_F \sigma_x k) \\ &\quad + Z^{-1}(\Lambda)(\omega_k - v_F \sigma_x k)\gamma(g) \log(\Lambda) \end{aligned} \quad (9)$$

where g is the effective coupling constant, given in terms of a suitable average value of the Coulomb potential \tilde{V} by $g = 2N_s N_T(\varepsilon_F) \tilde{V} / \pi v_F$. The function $\gamma(g)$ bears also an explicit dependence on the oscillating factor $N_T(\varepsilon_F)$, as it is given by

$$\gamma(g) = \frac{1}{4N_s N_T(\varepsilon_F)} \left(\sqrt{1+g} + \frac{1}{\sqrt{1+g}} - 2 \right) \quad (10)$$

The requirement of cutoff-independence of the renormalized Green function G is what forces the dependence of Z on the energy scale Λ , as it is bound then to satisfy the flow equation

$$\Lambda \frac{d}{d\Lambda} \log Z(\Lambda) = \gamma(g). \quad (11)$$

The quantity at the right-hand-side of (11) is actually the exponent α of the power-law behavior characterizing the suppression of the quasiparticle weight in the low-energy limit $\Lambda \rightarrow 0$. The novelty of our framework is that such an exponent depends now on the Fermi energy of the system, as the factor $N_T(\varepsilon_F)$ is sensitive to the position of the Fermi level in the structure of single-particle levels.

We now ascertain that our approach may account for the experimental features reported in Ref. 8. For this purpose, we have first to check that the Fermi energy shift $\Delta\varepsilon$ producing an oscillation in the α exponent is consistent with the variation of the gate voltage applied in the experiment. The dependence of α on the gate voltage V_g has been plotted in Figs. 2(c) and 2(d) of the mentioned reference. There it can be seen that, in a range of 20 V for the gate voltage, there are about eight oscillations in the α exponent. The corresponding change in the Fermi energy can be inferred from the capacitive coupling between the gate and the MWNT, that has been estimated to be about 1×10^3 [11]. Then, each oscillation in the plot should correspond to a variation in the Fermi energy of about 2 meV. This matches well the value of $\Delta\varepsilon$ estimated by taking the level spacing from the finite nanotube length L as a lower bound, $\Delta\varepsilon \sim h v_F / L \sim 1$ meV,

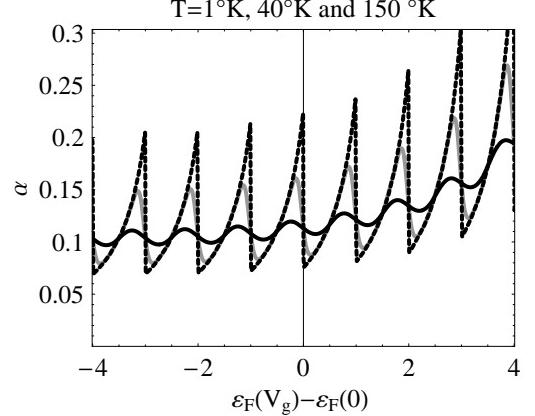


FIG. 2: Plot of the exponent α as a function of the Fermi energy ε_F (in units of $\Delta\varepsilon$) for different values of the temperature $T = 1, 40, 150$ K.

hence providing a strong indication that the quantization of the nanotube levels is at the origin of the oscillations observed in the α exponent.

The power-law behavior obtained from (11) applies directly to the TDOS $n(\varepsilon)$, since we have

$$n(\varepsilon) \sim N_s Z(\varepsilon) \sim \varepsilon^{\gamma(g)}. \quad (12)$$

We can compare the dependence of the exponent $\gamma(g)$ on ε_F with that of the power-law behavior of the conductance measured in Ref. 8. In our framework, the exponent depends on the position of the Fermi level due to the modulation introduced by $N_T(\varepsilon_F)$ in $\gamma(g)$, but also because of the possible variation of the Fermi velocity v_F as the Fermi level is shifted. In this respect, we recall that MWNTs are usually hole-doped, with several subbands crossing the Fermi level[12], so that a significant decrease in the Fermi velocity at some of the Fermi points is likely as the top of the lowest subband is approached. Actually, a drift in the average value of the exponent α is appreciable in Figs. 2(c) and 2(d) of Ref. 8, as the gate voltage runs from -10 V to 10 V. We have incorporated this effect by taking into account a suitable shift of the Fermi velocity in our model, from v_F to $5v_F/6$, as ε_F runs over 10 oscillations in the exponent. Finally, we have taken for the interaction strength $\tilde{V} \approx 5v_F$, bearing in mind the screening from the metallic shells of the MWNT[13]. The corresponding plot for α as a function of ε_F is given in Fig. 2. We observe the similarity of the plot with the experimental curves of Ref. 8 for the exponent over the 20 V range in the gate voltage.

Our approach provides also information about the dependence of the exponent α on the temperature. It can be seen in Fig. 2 that the oscillations are significant for $k_B T$ lower than a threshold of the order of $\Delta\varepsilon$. For higher temperatures, the thermal fluctuations erase the structure of superposed peaks in (2), what leads in turn to the disappearance of the oscillations in the α exponent.

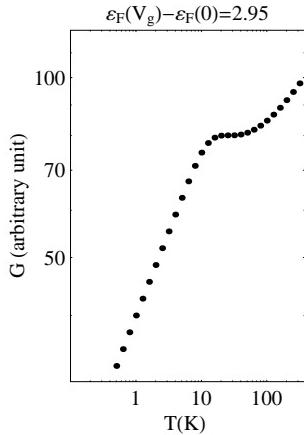


FIG. 3: Log-log plot of the conductance as a function of the temperature.

This agrees with the experimental observations, which show the existence of an inflection point T^* in the log-log plots of conductance versus temperature, for values of V_g corresponding to peaks in the plot of α . As reported in Ref. 8, the conductance keeps following a power-law behavior above the temperature T^* , but with an exponent significantly reduced with respect to the value at the peak.

We have checked that, for values of ϵ_F corresponding to peaks in the plot of α , an inflection point actually exists in the log-log plot of the conductance. This is obtained in our framework from the TDOS, by trading the energy variable in $n(\epsilon)$ for the corresponding thermal energy $k_B T$. The resulting dependence on temperature has been represented in Fig. 3. The value of the crossover temperature corresponds to a thermal energy of the order of $\Delta\epsilon$, which is consistent with the value of $T^* \approx 30$ K that has been found in the experiment.

We have thus achieved a description that accounts for the experimental features in the zero-bias conductance reported in Ref. 8. The discussion carried out in that paper highlighted the difficulty of finding a theoretical framework encompassing both the α oscillations and the

existence of the crossover temperature in the conductance. We have shown, however, that it is possible to extend the Luttinger liquid description of 1D electron systems by incorporating the effects of the discrete single-particle spectrum, thus producing a picture that is relevant to the mentioned experimental instance. We have seen that the actual size of the period for the α oscillations can be explained in our approach, in terms of the level spacing in the quantized spectrum. The estimated amplitude of the oscillations is also consistent with the values measured experimentally. Our theoretical construction accounts quite naturally for the existence of a crossover in the zero-bias conductance, keeping track of its power-law behavior both below and above the crossover temperature.

To conclude, we have developed a picture that applies to an intermediate regime between that of Coulomb blockade and the Luttinger liquid behavior. In the case of a MWNT with a length of a few microns, for instance, the finite size of the system leads to a spacing between single-particle levels which is of order ~ 0.1 meV. Thus, for temperatures which are $\lesssim 1$ K we have the Coulomb blockade regime, with the characteristic oscillations in the conductance as the gate voltage is varied. For temperatures well above that value, the conductance shows a power-law suppression as a function of the temperature, typical of Luttinger liquid behavior. We have shown that the intermediate regime can be characterized by a power-law behavior of the conductance, but with an exponent that oscillates as a function of the gate voltage, in agreement with observations reported in Ref. 8. This reassures the robustness of the Luttinger liquid picture for the description of the CNs, down to the low-energy scales where the collective excitations of the electron system begin to fade away.

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